

APPENDIX B
WASTE FORM SCREENING

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Important Waste Form Properties

The disposal of high-level radioactive waste is planned to be achieved through the immobilized emplacement of the waste in a deep geologic repository designed to provide multiple barriers to the release of radionuclides to the environment. Current reference designs for geologic repositories include a requirement that the stabilized waste form provide one of the many barriers to the release of radionuclides. Waste form properties that contribute to this function as a barrier include:

- Low leachability — the ability of the waste form to resist chemical dissolution in natural aqueous environments. Natural groundwater could provide a means both to degrade the waste form and to transport dissolved radionuclides to humans.
- Mechanical stability — the ability of the waste form to resist mechanical dispersion and to limit the surface area exposed to leaching.
- Radiation stability — the ability of the waste form to resist chemical or structural degradation due to radioactive decay of its radionuclides.
- Thermal stability — the ability of the waste form to resist chemical and physical degradation during the period when significant decay heat is generated in the waste.

Other waste form properties or characteristics important during production, handling, interim storage, shipment, repository emplacement, and repository retrieval (if required) are:

- Processing flexibility — the process must provide a consistent quality product over a range of operating parameters.
- Waste compatibility — the waste form must be able to accommodate the expected variations in waste composition.
- Mechanical strength — the waste form must resist thermal stress and the stress of normal handling.
- Impact resistance — the waste form must minimize the quantity of dispersible or respirable particles that would be produced by an impact accident.
- Fire resistance — the waste form must not release volatile radionuclides or generate gas which might rupture the canister during accidental external fires.

Finally, other waste form attributes could impact the costs of waste form production and disposal. These include:

- Process complexity - determines capital and operating expenses for waste form production.
- Waste loading - affects the number of waste canisters to be produced, packaged, shipped, and emplaced in the repository.

Candidate High-Level Waste Forms

The evaluation of potential waste forms for immobilization of SRP high-level waste began in 1973. In 1977, borosilicate glass was selected as the reference form for the DWPF. Since 1979, seventeen candidate materials (Table B-1), including borosilicate glass, have been considered as potential solid forms for the immobilization and geologic disposal of high-level waste. Screening evaluations^{1,2} during 1979 and 1980, based on performance potential and predicted process complexity of each form, reduced the number of forms from seventeen to seven. The evaluations considered nine scientific and nine engineering parameters affecting the long-term performance and production of waste forms. The elimination of ten of the forms from consideration was based upon such technical concerns as high porosities, high leach rates, questionable fracture behavior and tensile strength, incomplete partitioning of radionuclides within phases, possible effects of waste stream variation on phase assemblage and microstructure, potentially high corrosion rates, and potential phase sensitivity to radiation damage. Following continued development and characterization, the seven remaining forms (Table B-2) were evaluated further to select, in November 1981, two candidate forms for immobilizing SRP high-level waste.³

The selection of two of the seven forms for further development was based on four major inputs: (1) preliminary waste form evaluations conducted by the DOE defense waste sites for defense high-level waste and by an independent laboratory for commercial high-level waste; (2) peer review assessments and recommendations; (3) an evaluation of waste form product performance; and (4) an evaluation of waste form processability. The next two sections discuss the four major inputs considered in evaluating the seven candidate waste forms and the selection of the final two waste forms.

TABLE B-1

Candidate Waste Forms Considered for Geologic Disposal
of High-Level Waste

<u>Waste Form</u>	<u>Developer/Contractor</u>
Borosilicate Glass	Pacific Northwest Laboratory Savannah River Laboratory
High-Silica Glass	Catholic University of America NPD Nuclear Systems, Inc.
Phosphate Glass	Pacific Northwest Laboratory Brookhaven National Laboratory
Clay Ceramic	Rockwell Hanford Operations Pacific Northwest Laboratory
Glass Ceramic	Idaho Chemical Processing Plant
Tailored Ceramic	Rockwell International Pennsylvania State University
Synroc	Lawrence Livermore National Laboratory Argonne National Laboratory North Carolina State University
Titanate Ion Exchanger	Sandia National Laboratories
Stabilized Calcine	Idaho Chemical Processing Plant
Pelletized Calcine	Idaho Chemical Processing Plant
Normal Concrete	Pennsylvania State University Savannah River Laboratory Oak Ridge National Laboratory
Hot-Pressed Concrete	Pennsylvania State University
Concrete Formed Under Elevated Temperature and Pressure (FUEETAP)	Oak Ridge National Laboratory
Matrix Forms	Pacific Northwest Laboratory Argonne National Laboratory
Coated Sol-Gel Spheres	Oak Ridge National Laboratory
Cermet	Oak Ridge National Laboratory
Disc-Pelletized Coated Particles	Pacific Northwest Laboratory Battelle Columbus Laboratories

TABLE B-2

**Seven Candidate Waste Forms Evaluated for Geologic Disposal
of High-Level Waste**

<u>Waste Form</u>	<u>Developer/Contractor</u>
Borosilicate Glass	Pacific Northwest Laboratory Savannah River Laboratory
Synroc	Lawrence Livermore National Laboratory Argonne National Laboratory North Carolina State University
Tailored Ceramic	Rockwell International Pennsylvania State University
High-Silica Glass	Catholic University of America NPD Nuclear Systems, Inc.
Concrete Formed Under Elevated Temperature and Pressure	Oak Ridge National Laboratory
Coated Sol-Gel Spheres	Oak Ridge National Laboratory
Glass Marbles in a Lead Matrix	Pacific Northwest Laboratory

Screening Process

The first input considered in the evaluation of potential waste forms for immobilization of SRP high-level waste was a series of preliminary product and process evaluations⁴⁻⁷ conducted by each of the DOE defense sites (Savannah River, Hanford, and Idaho) to determine the preferred forms for immobilization of the high-level waste existing at each specific site. Additionally, two studies^{8,9} were conducted by Pacific Northwest Laboratory (PNL) to assess potential commercial waste forms and processes. Borosilicate glass was consistently the highest ranked form in each evaluation for immobilizing both defense and commercial high-level waste.* Either ceramic forms or other glass forms were the second most-preferred forms.

* In this discussion, rank and rate have the following meanings:
rank is used in the sense of an ordinal number giving relative standing or position;

rate is used to refer to a numerical value obtained through an evaluation or grading process.

As a second input, an Alternative Waste Form Peer Review Panel has been convened annually^{1,2} since 1979 to review the relative scientific merits and engineering practicality of high-level waste forms being developed. The panel's most recent review¹⁰ in May 1981 produced a relative ranking of the seven candidate forms. Borosilicate glass was ranked as the preferred form for immobilization of high-level waste followed in order by Synroc, high-silica glass, tailored ceramic, coated particles, FUETAP concrete, and glass marbles in a lead matrix.

A quantitative evaluation of waste form performance, the third input, was performed by the Savannah River Laboratory (SRL) using a rating system similar to one developed by a DOE Interface Working Group on High-Level Waste Form Selection Factors.¹¹ The evaluation compared the seven candidate waste forms on the basis of waste loading, mechanical stability, and leach resistance, with leach resistance given the highest weight. Waste loading was defined as curie content of SRP high-level waste per unit volume of waste form; mechanical stability was inferred from standard impact tests at Argonne National Laboratory; and leach resistance was determined by use of standard leach testing procedures developed by the Materials Characterization Center (MCC). Leaching data were provided by the waste form developers, MCC, and SRL.

Based on this evaluation,³ the waste forms were divided into three groups: (1) Synroc, tailored ceramic, and coated particles had the highest ratings; (2) borosilicate glass and high-silica glass had intermediate ratings; and (3) glass marbles in a lead matrix and FUETAP concrete had the lowest ratings. A clear delineation based on product performance could be made between the highest and lowest rated waste forms; distinctions between waste forms in the high and intermediate categories were less clear. The ceramic forms rated highest because they had the lowest uranium leach rates (the highest weighted single property); however, the glass forms rated better than the ceramics when considering leach rates for cesium and strontium (the main contributors to the curie content of the waste). Delineation among waste forms within a particular group was not possible based on product performance alone.

The fourth input was a processability analysis conducted by the Engineering Department of E. I. du Pont de Nemours and Company.¹² To evaluate quantitatively the waste form processes, twenty-one processability criteria were developed in four major categories: reliability/complexity, resource requirements, personnel safety, and quality control. Process data evaluated against these criteria were obtained from process flowsheets, equipment definitions, and conceptual facility layouts developed in collaboration with SRL and each of the waste form developers. The ratings based on processability fell in four general groups:

borosilicate glass and FUETAP concrete, relatively simple; glass marbles in a lead matrix and high-silica glass, moderately complex; crystalline ceramics, complex; and coated sol-gel particles, very complex.

Waste form ratings from the product performance and processability evaluations were combined to obtain an overall ranking of the seven waste forms. The ranking in order of highest to lowest was: borosilicate glass, Synroc and tailored ceramic, high-silica glass, FUETAP concrete, coated particles, and glass marbles in a lead matrix. Generally waste forms with high product performance ratings had low processability ratings, and vice versa. Borosilicate glass achieved the highest overall ranking because it had the highest processability rating combined with an intermediate product rating. The two ceramic forms ranked second overall because their high product ratings compensated for their lower processability ratings.

Screening Results

Based on the results of each of the four major inputs discussed above, borosilicate glass and crystalline ceramic were selected in November 1981 for further development as potential waste forms for immobilization of SRP high-level waste.

Borosilicate glass was selected for continued development on the following bases:

- Borosilicate glass demonstrated acceptable product performance properties.
- Borosilicate glass was ranked as the preferred form for high-level waste immobilization by the Alternative Waste Form Peer Review Panel.
- Borosilicate glass was consistently selected as the preferred form by the DOE defense sites, and was rated highest in the commercial waste form evaluations.
- The process for fabricating the borosilicate glass waste form is the simplest and least expensive of all those considered.

The crystalline ceramic forms, although ranking rather low in processing, were selected as the best alternative to borosilicate glass on the following bases:

- The crystalline ceramic forms, Synroc and tailored ceramic, ranked highest in the product performance evaluation.

- The Synroc form, ranked second by the Alternative Waste Form Peer Review Panel, was judged to be the best characterized and understood of the forms other than borosilicate glass.
- Ceramic waste forms consistently ranked high in each of the DOE defense-site evaluations.
- The ceramics have generally better high-temperature leaching characteristics than borosilicate glass.
- A number of mineral analogues of the crystalline ceramics have proven extremely durable in nature.

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